

FORM PTO-1390 (Modified) (REV 11-98)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER <b>TPP 30852</b>
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR <b>09/463560</b> )
INTERNATIONAL APPLICATION NO. <b>PCT/SE98/01309</b>	INTERNATIONAL FILING DATE <b>3 July 1998</b>	PRIORITY DATE CLAIMED <b>31 July 1997</b>	
TITLE OF INVENTION <b>A METHOD OF COATING A SUBSTRATE</b>			
APPLICANT(S) FOR DO/EO/US <b>LAMBERT, Nigel; STRACHAN, Adrian; WALLIS, Roger; and WRIGHT, Vincent</b>			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</li> <li>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li>6. <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</li> <li>7. <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210).</li> <li>8. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> have been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input checked="" type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>9. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li> <li>10. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).</li> <li>11. <input checked="" type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409).</li> <li>12. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).</li> </ol>			
Items 13 to 20 below concern document(s) or information included:			
<ol style="list-style-type: none"> <li>13. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li> <li>14. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li>15. <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment.</li> <li>16. <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</li> <li>17. <input type="checkbox"/> A substitute specification.</li> <li>18. <input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li>19. <input type="checkbox"/> Certificate of Mailing by Express Mail</li> <li>20. <input checked="" type="checkbox"/> Other items or information: <div style="border: 1px solid black; padding: 5px; margin-top: 5px;"> <ol style="list-style-type: none"> <li>a.) PCT/RO/101</li> <li>b.) PCT/IB/304</li> <li>c.) Copy of International Application as Published (WO 99/06489)</li> <li>d.) PCT/IPEA/402</li> <li>e.) PCT/IB/308</li> </ol> </div> </li> </ol>			

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.53) <div style="font-size: 24pt; font-weight: bold; margin-top: 5px;">09/463560</div>		INTERNATIONAL APPLICATION NO. <div style="font-weight: bold; margin-top: 5px;">PCT/SE98/01309</div>		ATTORNEY'S DOCKET NUMBER <div style="font-weight: bold; margin-top: 5px;">TPP 30852</div>	
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21. The following fees are submitted: <b>BASIC NATIONAL FEE ( 37 CFR 1.492 (a) (1) - (5) ) :</b> <div style="margin-top: 5px;"> <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... \$970.00  <input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... \$840.00  <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$690.00  <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$670.00  <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$96.00         </div> <div style="text-align: right; margin-top: 5px;"> <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b> </div>				<b>CALCULATIONS PTO USE ONLY</b>																																																																				
<div style="margin-top: 10px;">           Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).         </div> <table border="1" style="width: 100%; border-collapse: collapse; margin-top: 5px;"> <tr> <th style="width: 15%;">CLAIMS</th> <th style="width: 25%;">NUMBER FILED</th> <th style="width: 25%;">NUMBER EXTRA</th> <th style="width: 15%;">RATE</th> <th style="width: 20%;"></th> </tr> <tr> <td>Total claims</td> <td>22 - 20 =</td> <td>2</td> <td>x \$18.00</td> <td>\$36.00</td> </tr> <tr> <td>Independent claims</td> <td>1 - 3 =</td> <td>0</td> <td>x \$78.00</td> <td>\$0.00</td> </tr> <tr> <td colspan="4">Multiple Dependent Claims (check if applicable). <input type="checkbox"/></td> <td>\$0.00</td> </tr> <tr> <td colspan="4" style="text-align: right;"><b>TOTAL OF ABOVE CALCULATIONS =</b></td> <td>\$1,006.00</td> </tr> <tr> <td colspan="4">Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable). <input type="checkbox"/></td> <td>\$0.00</td> </tr> <tr> <td colspan="4" style="text-align: right;"><b>SUBTOTAL =</b></td> <td>\$1,006.00</td> </tr> <tr> <td colspan="4">Processing fee of <b>\$130.00</b> for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).</td> <td>\$0.00</td> </tr> <tr> <td colspan="4" style="text-align: right;"><b>TOTAL NATIONAL FEE =</b></td> <td>\$1,006.00</td> </tr> <tr> <td colspan="4">Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). <input type="checkbox"/></td> <td>\$0.00</td> </tr> <tr> <td colspan="4" style="text-align: right;"><b>TOTAL FEES ENCLOSED =</b></td> <td>\$1,006.00</td> </tr> <tr> <td colspan="4" rowspan="2"></td> <td style="text-align: center;">Amount to be:</td> <td>\$</td> </tr> <tr> <td style="text-align: center;">refunded</td> <td>\$</td> </tr> <tr> <td colspan="4" rowspan="2"></td> <td style="text-align: center;">charged</td> <td>\$</td> </tr> </table>				CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		Total claims	22 - 20 =	2	x \$18.00	\$36.00	Independent claims	1 - 3 =	0	x \$78.00	\$0.00	Multiple Dependent Claims (check if applicable). <input type="checkbox"/>				\$0.00	<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$1,006.00	Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable). <input type="checkbox"/>				\$0.00	<b>SUBTOTAL =</b>				\$1,006.00	Processing fee of <b>\$130.00</b> for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).				\$0.00	<b>TOTAL NATIONAL FEE =</b>				\$1,006.00	Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). <input type="checkbox"/>				\$0.00	<b>TOTAL FEES ENCLOSED =</b>				\$1,006.00					Amount to be:	\$	refunded	\$					charged	\$
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☒ A check in the amount of **\$1,006.00** to cover the above fees is enclosed.  
  
☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \_\_\_\_\_ to cover the above fees.  
 A duplicate copy of this sheet is enclosed.  
  
☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **19-4375** A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

**Thomas P. Pavelko, Esquire**  
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SIGNATURE

**Thomas P. Pavelko**

NAME

**31,689**

REGISTRATION NUMBER

**January 27, 2000**

DATE

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Sven KORNFALT et al

Attn: PCT Branch

Serial No.: National Stage Application based on  
International Application No. PCT/SE98/01309  
filed July 3, 1998

Filed: January 27, 2000

For: A METHOD OF COATING A SUBSTRATE

**PRELIMINARY AMENDMENT**

Honorable Commissioner of  
Patents and Trademarks  
Washington, D.C. 20036

Dear Sir:

Prior to an examination on the merits and calculation of the filing fee, please amend the above-identified application as follows:

**IN THE ABSTRACT**

Please substitute the attached one page Abstract with the one originally filed with the application.

**IN THE CLAIMS**

Claim 4, line 1, change "any one of the preceding Claims" to read --Claim 1--.

Claim 6, line 1, change "any one of the preceding Claims" to read --Claim 1--.

Claim 7, line 1, change "any one of the preceding Claims" to read --Claim 1--.

Claim 8, line 1, change "any one of the preceding Claims" to read --Claim 1--.

Claim 9, line 1, delete "7 or 8".

Claim 11, line 1, delete "or 10".

Claim 12, line 1, change "any one of the preceding Claims" to read --Claim 1--.

Claim 16, line 1, change "any one of the preceding Claims" to read --Claim 1--.

Claim 18, line 1, change "any one of the preceding Claims" to read --Claim 1--.

Claim 20, line 1, change "any one of the preceding Claims" to read --Claim 1--.

Claim 21, line 1, change "any one of Claims 1 to 19" to read --Claim 1--.

Claim 22, lines 1-2, change "any one of the preceding Claims" to read --Claim 1--.

Please cancel claim 23 without prejudice or disclaimer.

### REMARKS

The purpose of the foregoing Amendment is to delete multiple dependent claims, thereby minimizing the filing fee and placing the application in better form for examination under U.S. practice.

A prompt examination of the application is respectfully requested.

Respectfully submitted,



Thomas P. Pavelko  
Registration No. 31,689

TPP:mat  
Attorney Docket No.: TPP 30852

STEVENS, DAVIS, MILLER & MOSHER, L.L.P.  
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Telephone: (202) 785-0100  
Facsimile: (202) 408-5200 or (202) 408-5088

Date: January 27, 2000

THE APPLICANT HEREWITH PETITIONS  
THE PTO TO EXTEND THE TIME FOR  
RESPONSE TO BE REQUIRED TO MAKE THE  
ATTACHED DOCUMENT TIMELY FILED  
PLEASE CHARGE THE COST THEREOF  
TO DEPOSIT ACCOUNT 19-4373

STEVENS DAVIS MILLER & MOSHER, L.L.P.

Figure 1. The effect of the concentration of the *Agrobacterium* strain on the transformation efficiency of *Agrobacterium* strain 101. The concentration of the *Agrobacterium* strain 101 was varied from 10<sup>5</sup> to 10<sup>8</sup> cells/ml. The transformation efficiency was determined by the number of transformants per 10<sup>5</sup> cells of the *Agrobacterium* strain 101. The data are the mean  $\pm$  SD of three independent experiments. The transformation efficiency was significantly higher at 10<sup>7</sup> cells/ml than at 10<sup>5</sup> and 10<sup>6</sup> cells/ml ( $P < 0.05$ ).

[illegible]

420 Rec'd PCT/PTO 27 JAN 2000

A METHOD OF COATING A SUBSTRATE

The present invention relates to a method of coating a substrate.

It has been proposed previously to provide various coatings which can be applied to a substrate in a liquid form and which can then be cured to form a solid coating. Typically, the liquid coating incorporates unsaturated organic compounds which include C=C double bonds. These compounds present within the liquid coating are effectively polymerised during the curing process.

It has been proposed to effect the curing utilising high energy electron radiation. Typically electrons are delivered by an electron beam accelerator which normally operates at a voltage in excess of 150kVe, although alternatively a nuclear source may be utilised. The radiation breaks some of the C=C double bonds present in the unsaturated organic material, generating free radicals which initiate free radical polymerisation of the remaining material. The equipment necessary to carry out this process is costly to purchase and has to be specially shielded to avoid any leakage of gamma radiation.

There have been many proposals concerning coatings which can be cured, in response to ultra-violet light, involving free radical initiated polymerisation. Typically these coatings utilise a photo-initiator. A photo-initiator in this process is a material that absorbs light,

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and generates free radicals. The free radicals initiate the polymerisation of the coating. Photo-initiators are generally expensive, and can give rise to problems. For example, they can create an undesirable odour or taste (which may be relevant when the coating is going to be in contact with a food product) and can also cause "yellowing", which is a tendency for the cured coating to adopt a yellow colour over the course of time.

It has been proposed to provide a coating which can be cured on exposure to ultra-violet light which does not incorporate a photo-initiator. A coating of this type is disclosed in US-A-5,446,073. This Specification teaches a formulation which has a balance of "acceptor" and "donor" species. The process described in US-A-5,446,073 has not yet been adopted by industry, primarily because the reactive materials required are not readily commercially available. Also, the curing process is relatively slow with cure times typically being measured in terms of minutes. A further disadvantage of this technique is that it requires a combination of electron donating monomers and electron accepting monomers of relatively low molecular weight, and monomers in general are regarded as being prone to shrinkage during cure and are also regarded as being toxic since they may relatively easily penetrate the skin.

It has been discovered that short wavelength light may be used to effect a cure by direct fragmentation, in a similar way to the electron beam accelerator. Thus, it has been proposed to use light from excimer lamps, which have a wavelength of 172 nm, to cure radiation curable coatings without the use of a photo-initiator. However, this technique has only been used successfully with very thin coatings, typically coatings less than 1  $\mu\text{m}$  thick. The excimer energy is not able to penetrate readily into a

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coating which is of a greater thickness without excessive heat being generated. If an excimer lamp were used to irradiate a 10  $\mu\text{m}$  thick coating, it would produce a "cured skin" on the surface, but not a complete cure.

The present invention seeks to provide a UV cured coating in which the disadvantages of prior proposals are obviated or reduced.

According to one aspect of the present invention, there is provided a method of coating a substrate, the method comprising the steps of applying a coating composition to at least selected areas of the substrate, exposing the coated substrate to ultra-violet light from at least one lamp having a power output of at least 140 watts per linear centimetre in a curing zone, to initiate curing of the coating, the coating composition comprising a mixture including at least a reactive part comprising between 30% and 100% multi-functional material and being photo-initiator-free, including the step of maintaining a substantially inert atmosphere in the curing zone where the substrate is exposed to said ultra-violet light.

The preferred multi-functional materials have a functionality of at least three.

Preferably, the inert atmosphere is obtained by purging the curing zone with inert gas such as nitrogen.

Advantageously, the oxygen concentration in the curing zone is less than 1,000 ppm and preferably less than 100 ppm.

Preferably, the multi-functional material comprises one or more reactive diluents.



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Conveniently, the multi-functional material comprises one or more materials, each material having a molecular weight in excess of 480.

Advantageously, the multi-functional material comprises one or more materials which have three or more functional acrylate groups.

Conveniently, the coating material contains a pre-polymer, and may comprise polyester acrylate, polyurethane acrylate, epoxyacrylate or a full acrylic material.

Conveniently, the pre-polymer is multi-functional.

Advantageously, the coating composition comprises, in addition to the reactive part, a filler, and the filler may comprises clay, silica or magnetisable particles.

Preferably, the power output of the lamp is at least 180 watts/cm and may be substantially 240 watts/cm.

Conveniently, UV light from the lamp has a substantial spectral content in the range 200-300 nm.

Preferably, UV light from the lamp has additional spectral content with peaks of approximately 370 nm, 408 nm and 438 nm.

Two or more lamps may be provided in the curing zone. The lamps may have different spectral properties or may have substantially identical spectral properties.

The invention relates to a substrate when coated by a

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method as described above.

In order that the invention may be more readily understood, and so that further features thereof may be appreciated, the invention will now be described, by way of example, with reference to the accompanying drawings in which:

FIGURE 1 is a diagrammatic view of an apparatus for use in curing a coating on a substrate;

FIGURE 2 is a graphical indication of the spectral output of a preferred UV lamp;

FIGURE 3 is a graphical representation of the spectral output of an alternative preferred UV lamp; and

FIGURE 4 shows the chemical structure of a reactant.

Referring initially to Figure 1 of the accompanying drawings, an apparatus for curing a coating applied to a substrate is illustrated.

An apparatus is illustrated which comprises a pair of rollers 1,2 adapted to guide a substrate 3, such as a sheet of aluminium foil or a sheet of paper, through the illustrated apparatus. The substrate 3 is coated, before entering the apparatus, completely or partially, with a curable composition which will be described in greater detail hereinafter. The curable composition may be applied as an un-broken coating or may be applied in the form of printing.

The rollers 1,2 guide the substrate 3 through a channel 4 defined between a cooled backing plate 5, and a

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super-imposed cover 6. The cover 6 supports two lamps 7,8.

The lamp 7 can be a "D" lamp, as supplied by Fusion Inc. The lamp 7 is located above a quartz window 9 that is sealed against the cover 6 so that light from the lamp 7 may shine through the quartz window 9 on to the substrate 3 as it passes through a curing zone formed by the channel 4.

The lamp 8 can be a "H" lamp as supplied by Fusion Inc. and is also associated with a quartz window 10 which is formed integrally with the cover 6 so that light from the lamp 8 can pass through the quartz window 10 into the curing zone in channel 4, and thus on to the substrate 3.

The lamps 7,8 each have a focussing reflector. The position of the lamps may be interchanged. In an alternative embodiment, only one lamp may be used, or two lamps of the same type may be used.

The lamps 7 and 8 emit UV light with wavelengths in the band 200 to 550 nm. The light is directed on to the substrate in a region in the central part of the channel 4.

The channel 4 is flushed with nitrogen in order to ensure that there is an inert atmosphere having a minimum quantity of oxygen within the channel. Thus, nitrogen is introduced through an inlet conduit 11 and passes to a dispersing nozzle 12 known as the "inlet knife" which is located on the under-side of the cover 6 above the roller 1, and which is adapted to prevent oxygen entering the channel 4. The nitrogen flows along the channel 4, past the region where the UV light is directed on to the substrate, in the same direction as the direction of movement of the substrate 3. Nitrogen may also be injected into the channel 4 via nozzles located around the periphery

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of each quartz window. At the end of the channel 4 adjacent the roller 2 nitrogen flowing through a conduit 13 is passed through a nozzle 14 on the underside of the cover into the channel 4. Thus, there is a continuous flow of nitrogen within the channel 4 such that the concentration of oxygen within the channel 4 is less than 1,000 ppm and preferably less than 100 ppm. Instead of using nitrogen, other inert gases could be used. However, nitrogen is preferred because it is the least expensive inert gas.

The lamps 7 and 8 are supplied in a modular form, each module is 25.4 mm wide and extends transversely across the direction of movement of the web 3 through the channel 4. The lamps are high intensity lamps using medium pressure mercury vapour bulbs operating at a power level in excess of 140 watts per linear centimetre, preferably in excess of 180 watts per linear centimetre, and most preferably in the region of 240 watts per linear centimetre.

The output spectrum of the "D" lamp 7 is illustrated in Figure 2. It can be observed that the spectrum has a substantial spectral content within the range of 350-450 nm, with specific peaks at 355, 370, 383 and 408 nm. There is spectral content in the region of 200-350 nm, although the spectral content in the 200-250 nm range is minimal.

The spectrum of the "H" lamp 8 is illustrated in Figure 3. It can be seen that the spectrum has substantial spectral content in the region of 200-300 nm, with the spectral content at the longer wavelengths being restricted to isolated peaks at 312 nm, 370 nm, 408 nm and 438 nm. The spectral content in the region of 200-300 nm comprises a very broad peak centred on 225 nm and spanning the area from 210-240 nm and a further relatively broad peak, which is less clearly defined, but which is substantially centred

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on 262 nm and effectively spans the range of 240-280 nm. It is believed that this very substantial spectral content at these very short wavelengths plays a significant role in creating free radicals to initiate polymerisation. The very high energy present in the well defined peaks at higher frequencies may also contribute.

The reactive part of the curable coating that is applied to the web 3 does not contain a photo-initiator, but does contain a substantial proportion (between 30% and 100% by weight) of multi-functional radiation curable elements. A multi-functional radiation curable element is a radiation curable element which comprises two or more functional groups. Functional groups are acrylate groups with C=C double bonds. If functionality is expressed as a number, the number indicates the number of C=C double bonds available to react, present in acrylate groups.

The radiation curable elements are preferably of low viscosity and can be considered to be reactive diluents, not only providing reactive capabilities, but also maintaining, in the unreacted state, the desired liquid properties of the coating material.

It is preferred that the average molecular weight of any single multi-functional radiation curable element utilised in the curable coating should be greater than 480.

It has been found that relatively low molecular weight radiation curable elements may give rise to skin irritation. It is, however, believed that by utilising a molecular weight greater than 480, the risk of skin irritation arising is substantially reduced or obviated.

Typical examples of multi-functional radiation curable reactive diluents are propoxylated pentaerythritol tetra-

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acrylate or ethoxylated pentaerythritol tetra-acrylate. An alternative material comprises "OTA 480", a triacrylated low viscosity material available from UCB Chemicals of Anderlecht Str. 33, B-1620 Drogenbos, Belgium. The structure of OTA 480 is shown in Figure 4.

The curable coating may optionally comprise, in addition to the reactive part, a non-reactive part or filler that may comprise clay or silica. In some cases, where the coating is to have magnetic properties, the filler may comprise metal particles that may be magnetised.

It is believed that the very high intensity UV radiation applied by the lamps to the curable material generates sufficient free radicals to initiate the curing of the coating.

As the reaction takes place within the substantially inert atmosphere, it is thought that even though a relatively low number of radicals may be produced, because of the high functionality of the coating material, and because the radicals are not subject to oxygen quenching, the radicals that are available to initiate the reaction are sufficient to enable the reaction to proceed very rapidly. Although multi-functional materials, where functionality is greater than 3, are highly reactive, they are believed to undergo a relatively low level of conversion to form a fully cured coating, when compared to mono or di-functional materials.

It is believed that the physical properties of a coating formed solely from multi-functional reactive diluents of low viscosity, whilst sufficient for many purposes, may not be considered sufficient for use as a high performance coating. In order to produce a coating

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formulation that exhibits solvent resistance and stain resistance, it has been found appropriate to combine, with the reactive diluents, a pre-polymer which preferably contains some unsaturation. A pre-polymer is a reactive material of relatively high viscosity. A pre-polymer, when used alone, does not exhibit liquid properties that are appropriate for a coating material that could be used for example in a direct gravure coating process. Examples of suitable pre-polymers are polyester acrylates, polyurethane acrylates and epoxy acrylates. The functionality of these materials is normally 2 or 3 but can be up to 6. It is thought that the higher the functionality of the pre-polymers, the faster the curing performance of the coating.

#### EXAMPLE 1

A series of multi-functional materials were coated on to aluminium foil at a coat weight of approximately  $10\text{g/m}^2$ .

The coated foil was passed through a curing apparatus, similar to that shown in Figure 1, but with only one lamp at a speed of 20 m/min. Initially, the lamp was an "H" lamp, and subsequently the experiment was repeated using a "D" lamp. The lamps were each operated at a power level of 240 watts/centimetre. The cured coating was subsequently tested using an acetone rub test utilising a SATRA rub tester. Such a tester provides an indication of the degree of curing. Solvent resistance is indicated as the number of double rubs effected before the coating applied to the aluminium foil is removed. The greater the number of rubs, the more solvent resistant is the coating.

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The results are set out in Table 1.

TABLE 1

Coating Composition	Solvent Resistance - Acetone rubs	
	H Bulb	D Bulb
<u>Di and Triacrylates</u>		
Tripropan-glycol- diacrylate (UCB)	<2	<2
1,6-hexanediol diacry- late (UCB)	25	<2
OTA 480 triacrylate (UCB)	40	<2
<u>Tetra-acrylates</u>		
Ebecryl 40 (UCB)	95	<2
Ethoxylated- pentaerythritol tetra-acrylate (Croda)	45	<2
<u>Tetra-acrylate with pre-polymer</u>		
Actilane 320 PP 50 Epoxy acrylate with 50% Propoxylated- pentaerythritol tetra-acrylate (Ackros)	150	<2

Ebecryl 40 is a tetra-functional acrylated reactive diluent available from UCB Chemicals. The ethoxylated pentaerythritol tetra-acrylate was obtained from Croda Resins Ltd. of Crabtree Manorway, South Belvedere, Kent DA17 6BA. Actilane 320 PP 50 is obtained from Ackros Chemicals, Eccles Site, Bentcliffe Way, P.O. Box 1, Eccles, Manchester M30 0BH.



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Example 1 shows that when the coating composition comprises only a di-acrylate, the solvent resistance has a very low value. When tri-acrylates or tetra-acrylates are utilised, a much greater degree of solvent resistance is achieved. When a tetra-acrylate mixed with a pre-polymer is utilised, there is substantial improvement in the performance of the cured coating. This is believed to be due to the introduction of higher molecular weight epoxy groups in the cross-linked structure of the coating.

#### EXAMPLE 2

A coating of HH52-0103 02 EBC foil coating material obtained from Glasurit GmbH, Postfach 6123, D-48136 Muenster, Germany, was applied at a coat weight of approximately  $10\text{g/m}^2$  to an aluminium foil. This material comprises a relatively small proportion epoxy acrylate and a relatively large proportion of ethoxylated pentaerythritol tetra-acrylate. The coated foil was passed at various speeds through a curing apparatus similar to that shown in Figure 1 but with only a single "H" lamp 7 operating. In an initial series of experiments, the "H" lamp was operated at a power of 240 watts per centimetre and in another series of experiments the "H" lamp was operated at a power of 160 watts per centimetre. The acetone rub test as described with reference to Example 1, was subsequently used to indicate the degree of curing. The results are shown in Table 2.

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TABLE 2

Speed	Solvent Resistance Acetone rubs	
	@ 240 W/cm	@ 160 W/cm
30 M/min	100	11
40 M/min	35	9
50 M/min	9	1
60 M/min	4	1

Example 2 illustrates that the degree of curing of the coating is improved when the coating is provided with a substantial energy input in the UV spectrum. The best results are achieved with a relatively low speed of movement of the substrate through the curing apparatus and with the application of very intense high energy light. It can be seen that it is important, for a satisfactory cure to be achieved, for there to be a sufficient power input for a sufficient period of time. The degree of cure achieved does not increase linearly with lower speed of passage through the curing zone (i.e. does not increase linearly with the exposure time to intense UV light). As can be seen, especially at 240 watts per centimetre, the degree of cure achieved increases very substantially, almost exponentially, with increased residence time in the curing zone.

It is believed that an even higher degree of cure can be achieved utilising two lamps within the curing zone. This is shown in Example 3 where the same coating is applied and cured using two lamps at a much higher speed.

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EXAMPLE 3

A coating of HH52-0103 02 EBC foil coating material was coated on to a printed paper (Paper 13-2606), using a gravure coating unit at a coat weight of approximately 8 g/m<sup>2</sup> and a run speed of 80 metres per minute. The coated paper was passed through a UV curing apparatus of a type illustrated in Figure 1 flushed with nitrogen gas to as to achieve a residual oxygen level within the curing region of less than 100 ppm. The curing zone was illuminated, through quartz windows, by two high intensity UV lamps, one "H" bulb and one "D" bulb utilising focus-reflectors to direct UV light on to the coating to cure it to form a dry film, the two lamps operating at a power of 240 watts per linear centimetre.

The solvent resistance was greater than 50 acetone rubs (measured as in Example 2).

To test the performance of the coated paper as a furniture surfacing material, a sample of the coated paper was laminated, using aminoplast adhesive, on to a piece of chipboard. Then stain and scratch tests were carried out using the techniques described in ISO 4211. For comparison purposes, the test run was repeated, adding to the coating material the photo-initiator recommended by, and obtainable from, Glasurit GmbH, which is a methyl-phenyl-glyoxylate identified as SR07 641H. This coated paper was also laminated to a piece of chipboard using aminoplast adhesive. For further comparison, a sample of commercially available furniture foil which had been lacquered using a water-based amino polyester lacquer and then thermally



TABLE 3

Test Liquid	ISO 4211 Stain Resistance		
	Thermal Cured	UV No. Photoinitiator	UV + 0.5 % SR07-641H
	Lacquer		
Acetic Acid	3.0	3.0	2.0
Acetone	3.0	3.0	3.0
Ammonia solution	3.0	3.0	3.0
Blackcurrant juice	3.0	3.0	3.0
Citric acid	3.0	3.0	3.0
Cleansing agent	3.5	3.0	3.0
Coffee	3.0	3.5	3.5
Disinfectant (DETTOL)	3.5	4.0	4.0
Disinfectant (SAVLON)	3.0	4.0	4.0
Endorsing ink	4.0	3.0	3.0
Ethanol	3.5	3.5	3.5
Ethyl/Butyl acetate	3.5	3.5	3.0
Iodine	4.0	3.0	3.0
Milk (condensed)	3.0	3.0	3.0
Olive Oil	3.5	3.5	3.5
Paraffin oil BP	3.5	3.5	3.0
SBP Spirit	4.0	3.0	3.0
Sodium carbonate	4.0	4.0	4.0
Sodium chloride	4.0	4.0	3.0
Tea	4.0	4.0	3.5
Water	3.5	3.5	3.0
TOTALS:	72.5	71.0	67.0

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Example 3 indicates that a coating composition in accordance with the present invention, with no photo-initiator, provides a superior performance to a coating composition which does include a photo-initiator. The performance achieved by the material of the present invention is very similar to the performance achieved by the industry-acceptable-standard utilised for comparison purposes. Furthermore, by utilising a preferred combination of UV lamp power, spectrum and reactive materials, the process can operate at higher speeds.

Consequently, it is believed that the present invention provides a method of producing an industry-acceptable material without the use of photo-initiators, but whilst still providing the other advantages of UV curing.

The coating techniques described above have been found to be particularly suitable for applying coatings to flexible papers or films, such as papers or films produced on high speed coating and printing machines. The coatings have been found to be especially valuable for use on surfaces that are found in the home environment. Thus, the coatings may be applied to surfacing materials intended for application to furniture, walls, floors and ceilings. However, it is to be understood that the method described above may be utilised for less demanding coating applications, such as varnishes for books, magazines or record sleeves. The method described above may also be utilised for fabricating coated materials for use in packaging where low odour coatings, or coatings which do not impart a "taste" to products, are of particular value.

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CLAIMS:

1. A method of coating a substrate, the method comprising the steps of applying a coating composition to at least selected areas of the substrate, exposing the coated substrate to ultra-violet light from at least one lamp having a power output of at least 140 watts per linear centimetre in a curing zone, to initiate curing of the coating, the coating composition comprising a mixture including at least a reactive part comprising between 30% and 100% multi-functional material and being photo-initiator-free, including the step of maintaining a substantially inert atmosphere in the curing zone where the substrate is exposed to said ultra-violet light.
2. A method according to Claim 1 wherein the inert atmosphere is obtained by purging the said curing zone with inert gas.
3. A method according to Claim 2 wherein the inert gas comprises nitrogen.
4. A method according to any one of the preceding Claims wherein the oxygen concentration within the said curing zone is less than 1,000 parts per million.
5. A method according to Claim 4 wherein the oxygen concentration is less than 100 parts per million.
6. A method according to any one of the preceding Claims wherein the multi-functional material comprises one or more reactive diluents.
7. A method according to any one of the preceding Claims wherein the multi-functional material comprises one or more

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materials, the or each material having a molecular weight in excess of 480.

8. A method according to any one of the preceding Claims wherein the multi-functional material comprises one or more materials which have three or more functional acrylate groups.

9. A method according to Claim 6, 7 or 8 wherein the coating material additionally contains a pre-polymer.

10. A method according to Claim 9 wherein the pre-polymer comprises polyester acrylate, polyurethane acrylate, epoxyacrylate, or a full acrylate material.

11. A method according to Claim 9 or 10 wherein the pre-polymer is multi-functional.

12. A method according to any one of the preceding Claims wherein the coating composition comprises, in addition to the reactive part, a filler.

13. A method according to Claim 12 wherein the filler is clay.

14. A method according to Claim 12 wherein the filler is silica.

15. A method according to Claim 12 wherein the filler is magnetisable particles.

16. A method according to any one of the preceding Claims wherein the power output of the lamp is at least 180 watts/cm.



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17. A method according to Claim 16 wherein the power output of the lamp is substantially 240 watts/cm.

18. A method according to any one of the preceding Claims wherein UV light from the lamp has a substantial spectral content in the range of 200-300 nm.

19. A method according to Claim 18 wherein UV light from the lamp has a spectral content at peaks of approximately 370 nm, 408 nm and 438 nm.

20. A method according to any one of the preceding Claims wherein two lamps are provided in the curing zone, the lamps having different spectral properties.

21. A method according to any one of Claims 1 to 19 wherein two lamps are provided in the curing zone, the lamps having substantially identical spectral properties.

22. A substrate when coated by a method according to any one of the preceding Claims.

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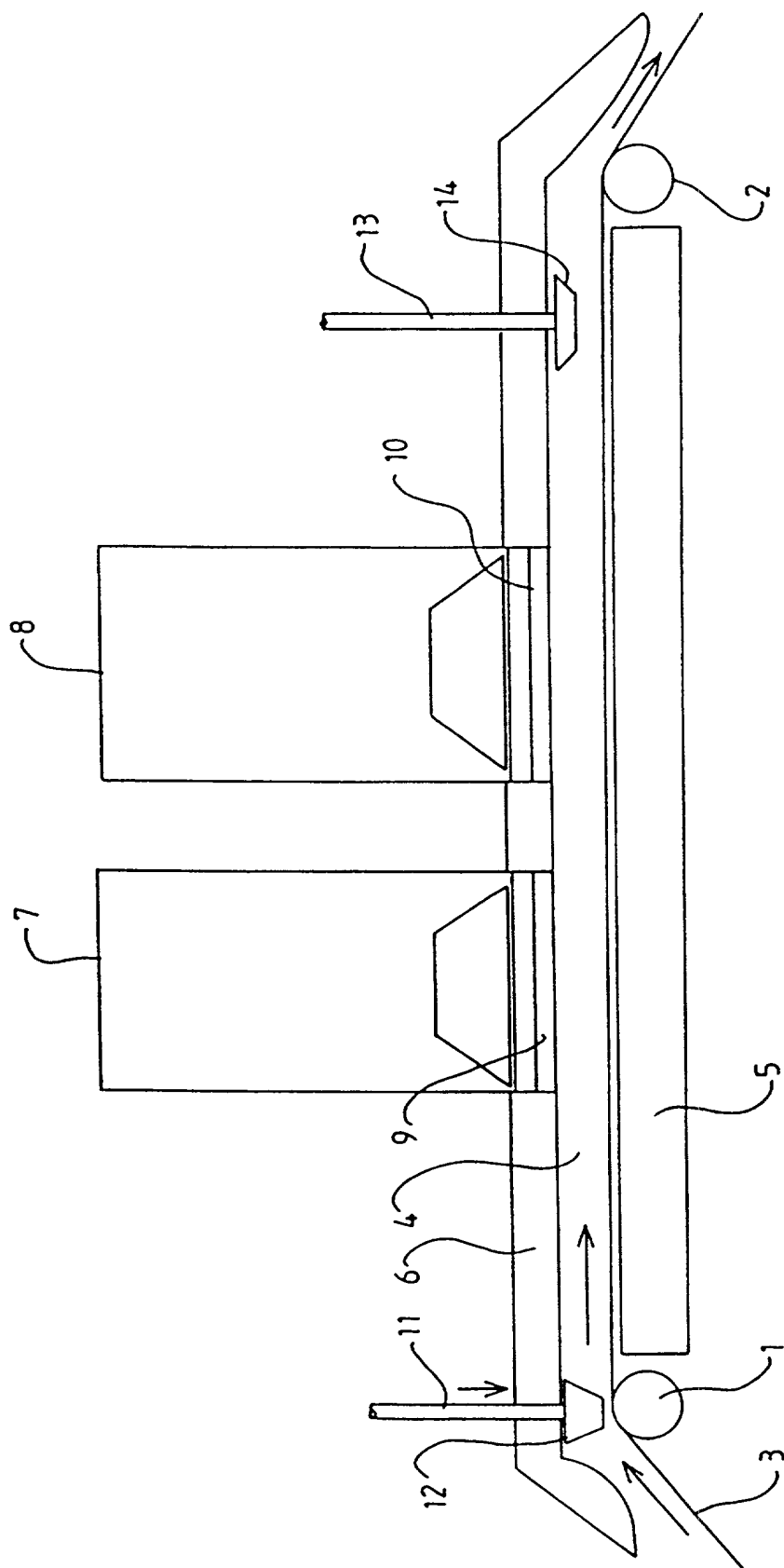
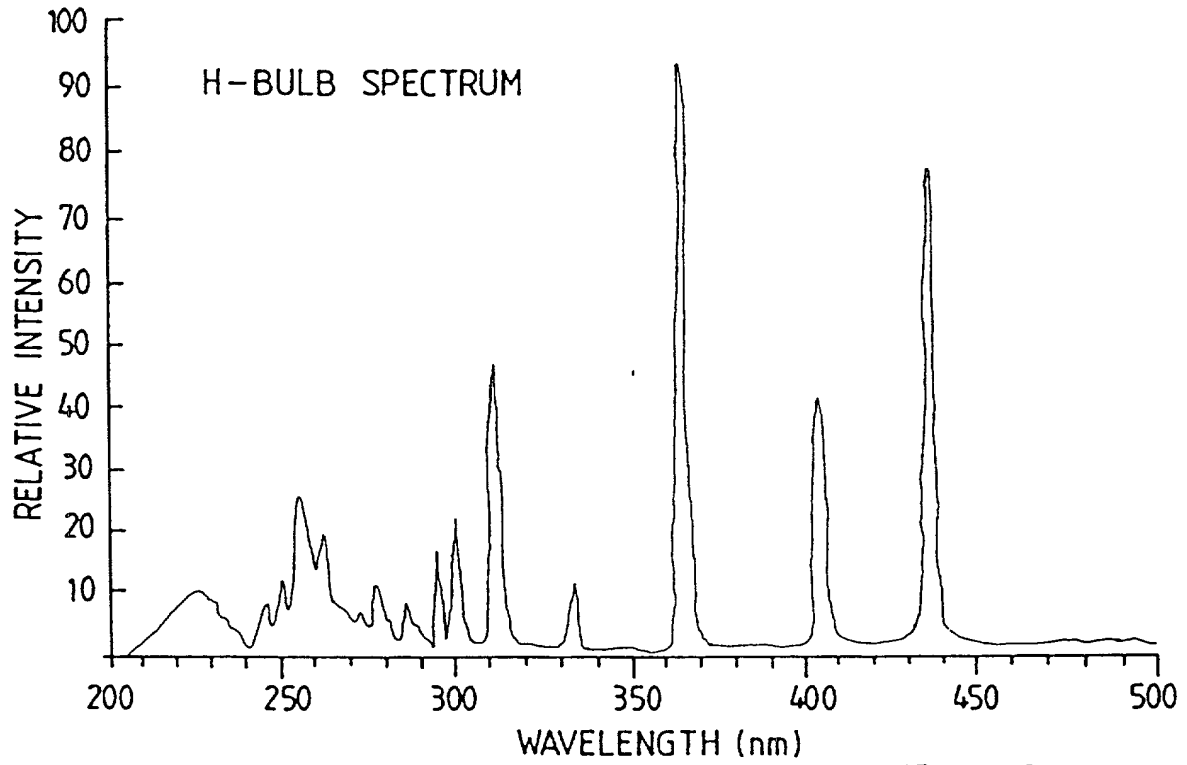
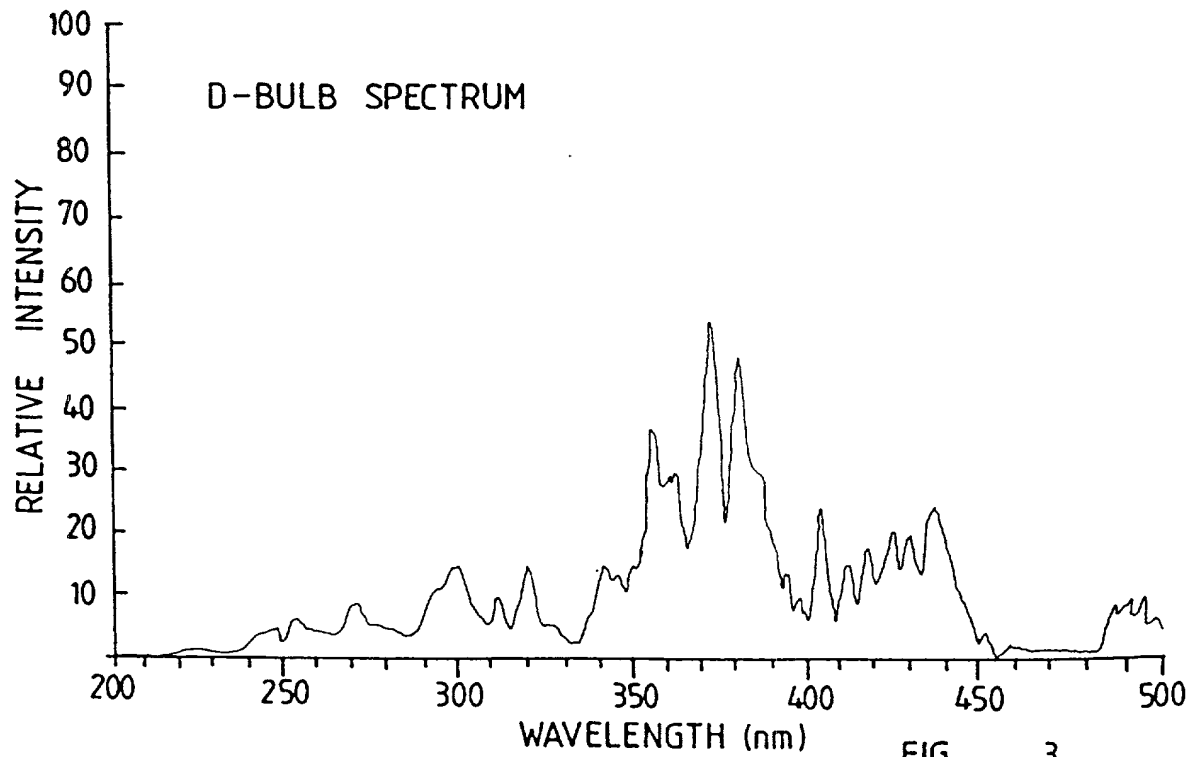
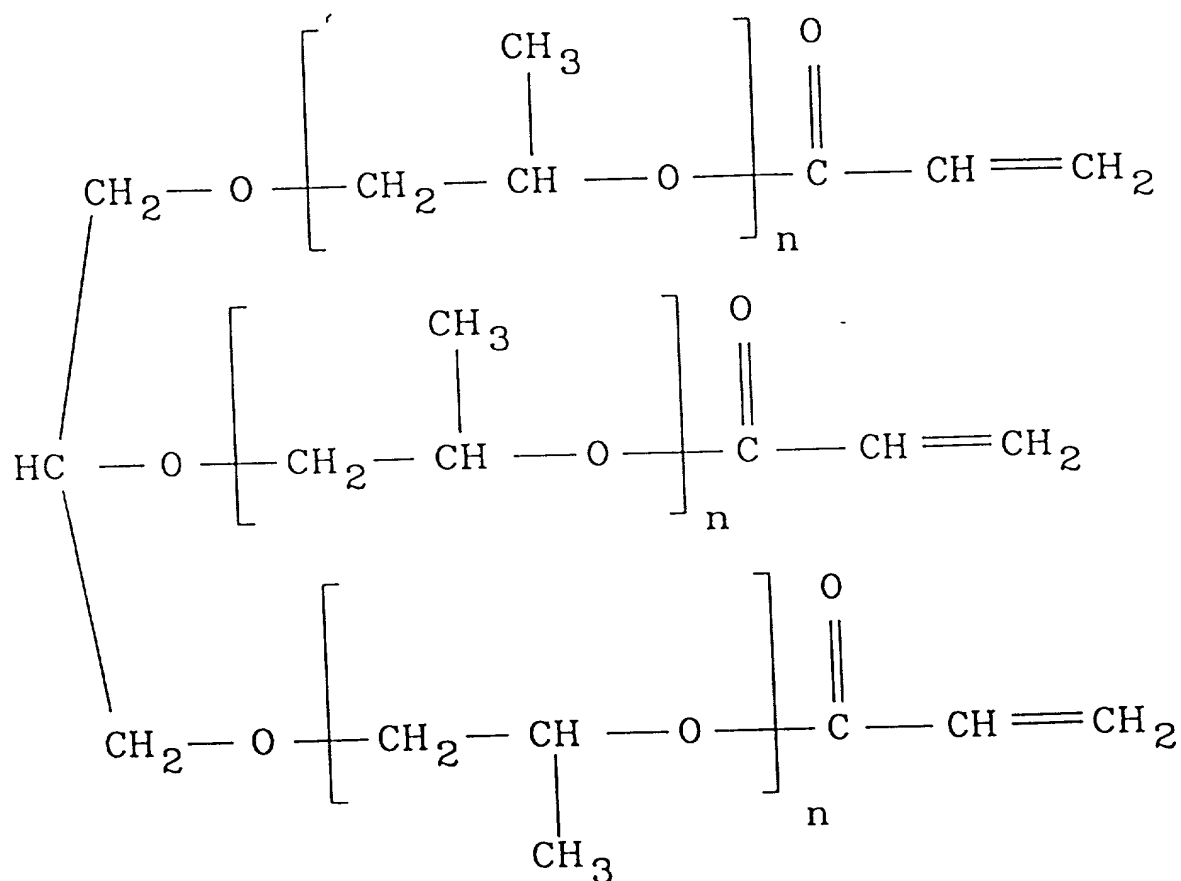


FIG 1

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FIG 2FIG 3

3 7 3

FIG 4

21 MAR 2000

09/463560 Case 581 215

COMBINED DECLARATION AND POWER OF ATTORNEY FOR  
UTILITY PATENT APPLICATION (Includes PCT)

Attorney Docket No.  
TPP 30852

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name;  
that

I believe I am the original, first and sole inventor (if only one name is listed below)  
or an original, first and joint inventor (if plural inventors are listed below) of the  
subject matter which is claimed and for which a patent is sought on the invention entitled:

A METHOD OF COATING A SUBSTRATE

the specification of which (check one)

☐ is attached hereto.

☐ was filed on \_\_\_\_\_ as Application Serial No. \_\_\_\_\_

and was amended on \_\_\_\_\_  
(if applicable)

☒ was filed as PCT international application no. PCT/SE98/01309

on 03 July 1998, and was amended under PCT Article 19

on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified  
specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this  
application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I do not know and do not believe the claimed invention was ever known or used in the United  
States of America before my or our invention thereof, or patented or described in any printed  
publication in any country before my or our invention thereof or more than one year prior to  
this application, that the same was not in public use or on sale in the United States of  
America more than one year prior to this application, that the invention has not been  
patented or made the subject of an inventor's certificate issued before the date of this  
application in any country foreign to the United States of America on an application filed  
by me or my legal representatives or assigns more than twelve months prior to this  
application.

I hereby claim foreign priority benefits under Title 35, United States Code §119 of any  
foreign application(s) and United States provisional applications for patent or inventor's  
certificate listed below and have also identified below any foreign application for patent  
or inventor's certificate having a filing date before that of the application(s) on which  
priority is claimed:

Prior Foreign and U.S. Provisional Application(s)

Priority Claimed

<u>9716194.7</u>	<u>England</u>	<u>31 July 1997</u>	<input checked="" type="checkbox"/>	<input type="checkbox"/>
(Number)	(Country)	Day/Month/Year Filed	Yes	No
_____	_____	_____	<input type="checkbox"/>	<input type="checkbox"/>
(Number)	(Country)	Day/Month/Year Filed	Yes	No

Attorney Docket No. TPP 30852

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

Application Serial No.	Filing Date	Status
		(patented, pending, abandoned)



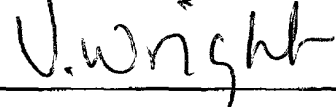
Application Serial No.	Filing Date	Status
		(patented, pending, abandoned)

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith; Stevens, Davis, Miller & Mosher, L.L.P.; Anthony P. Venturino, Reg. No. 31,674; James E. Ledbetter, Reg. No. 28,732; and Thomas P. Pavelko, Reg. No. 31,689. Direct all telephone calls to telephone no. (202) 785-0100 and faxes to (202) 202-408-5200.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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